

September 20, 2016

Dear Dr. Logan,

We have submitted a manuscript entitled “Legacy and emerging perfluoroalkyl substances are important drinking water contaminants in the Cape Fear River Watershed of North Carolina” as a research article for *Environmental Science & Technology Letters*. The total word count is 2960 including figures.

In our view, the submitted manuscript fills important knowledge gaps associated with the occurrence of emerging perfluoroalkyl substances (PFASs) and their behavior during water treatment. Because of the growing public health concerns associated with long-chain PFASs, fluorochemical manufacturers have replaced long-chain PFASs with short-chain PFASs and fluorinated alternatives. To date, very limited information is available about these alternatives. Thus, the goals of this study were to (1) determine the occurrence of legacy PFASs and emerging perfluoroalkyl ether carboxylic acids (PFECAs) in a watershed that serves as a drinking water source for 1.5 million people, (2) assess the behavior of PFECAs in conventional and advanced water treatment processes, and (3) identify PFECA characteristics that affect their adsorptive removal by activated carbon.

In this paper we found (1) high concentrations of legacy PFASs still occur in one drinking water source, even though their production has been phased out in the US; (2) PFECAs dominate the PFAS signature downstream of a PFAS production facility, illustrating the insufficiency of current analytical methods (EPA Method 537) and the need for updated monitoring approaches and discharge regulations; (3) removal of both legacy and emerging PFASs was negligible in a full-scale water treatment plant that employs state-of-the art water treatment processes, including ozonation, biofiltration, and UV disinfection; and (4) replacement of legacy PFASs with alternatives (e.g., replacing perfluorooctanoic acid with perfluoro-2-propoxypropanoic acid, a.k.a. “GenX”), a less absorbable compound is introduced into drinking water sources, making activated carbon adsorption a less feasible removal option. This manuscript highlights that communities served by public water systems are exposed to high levels of both legacy and emerging PFASs and highlights the need for new monitoring approaches and discharge regulations. We look forward to receiving editorial and peer review comments on this work. Thank you for your consideration of our manuscript.

Sincerely,

Mei Sun, Assistant Professor
University of North Carolina at Charlotte
Department of Civil and Environmental Engineering
9201 University City Blvd | Charlotte, NC 28223
Email: msun8@uncc.edu | Phone: 704-687-1723 | Fax: 704-687-0957

List of related work submitted or published

- Strynar, M.; Dagnino, S.; McMahan, R.; Liang, S.; Lindstrom, A.; Andersen, E.; McMillan, L.; Thurman, M.; Ferrer, I.; Ball, C., Identification of novel perfluoroalkyl ether carboxylic acids (PFECAs) and sulfonic acids (PFESAs) in natural waters using accurate mass time-of-flight mass spectrometry (TOFMS). *Environmental Science & Technology* 2015, 49, (19), 11622-11630.
- Dudley, L. A.; Arevalo, E. C.; Knappe, D. R. U. *Removal of perfluoroalkyl substances by PAC adsorption and anion exchange*; Water Research Foundation: Denver, CO, 2015.
- Nakayama, S.; Strynar, M. J.; Helfant, L.; Egeghy, P.; Ye, X.; Lindstrom, A. B., Perfluorinated compounds in the Cape Fear drainage basin in North Carolina. *Environmental Science & Technology* 2007, 41, (15), 5271-5276.

Suggested Reviewers:

Chris Higgins, Associate Professor
Colorado School of Mines
Department of Civil & Environmental Engineering
Email: chiggins@mines.edu
Tel: (303)-384-2002
<http://inside.mines.edu/Higgins>

Shubo Deng, Professor
Tsinghua University, China
Department of Environmental Science and Engineering
E-mail: dengshubo@tsinghua.edu.cn
Phone: 86-10-62792165
http://www.tsinghua.edu.cn/publish/enven/6309/2011/20110215133411785291121/20110215133411785291121_.html

Ralf Ebinghaus, Professor
Institute of Coastal Research, Germany
Department for Environmental Chemistry
E-mail: ralf.ebinghaus@hzg.de.
Phone: 49-4152-87-2354
http://www.hzg.de/institutes_platforms/coastal_research/biogeochemistry_in_coastal_seas/environmental_chemistry/staff/046361/index.php.en

Andy Eaton PhD, BCES
Technical Director, Eurofins Eaton Analytical Inc
750 Royal Oaks Drive
Monrovia, CA 91016 USA
Email: AndyEaton@eurofinsUS.com
Phone: 626-386-1125

Susan Richardson, Arthur Sease Williams Professor of Chemistry / Analytical
University of South Carolina
Department of Chemistry and Biochemistry
E-mail: richardson.susan@sc.edu
Phone: 803-777-6932
https://sc.edu/study/colleges_schools/chemistry_and_biochemistry/our_people/richardson_susan.php